

Origins of Conductive Losses at Microwave Frequencies in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}/\text{LaAlO}_3/\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ Trilayers Deposited by Pulsed Laser Deposition

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Oriented $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}/\text{LaAlO}_3/\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ trilayers were deposited by pulsed laser deposition (PLD) onto $\langle 100 \rangle$ MgO and LaAlO_3 . Film thicknesses varied from 2000–5000 Å/layer. A comparison of structure and transport data for the bottom and top superconducting layers indicated a slight decrease in film quality for the top superconducting layer. The critical temperature was lower for the top superconducting layer (90.5 vs. ~ 90 K) and the microwave surface resistance was higher (increasing from ~ 2 to 18 mΩ at 36 GHz, 20 K). The resistivity of the dielectric was estimated to be 10^6 Ω cm, and the loss tangent of the dielectric film at microwave frequencies had an upper limit of 0.01. Cross-sectional TEM analysis of the trilayer structure showed a high density of threading dislocations in the dielectric layer that appeared to nucleate at steps in the underlying superconducting layer. The threading dislocations may serve as conduction paths in the LaAlO_3 layer.

KEY WORDS: $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$; pulsed laser deposition; LaAlO_3 ; microwave surface resistance; thin film microstructure; TEM.

1. INTRODUCTION

The development of epitaxial thin-film insulators compatible with epitaxial $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) is important to the fabrication of low loss multilayer devices and circuit structures. Insulating materials are needed for junction barriers, cross-overs, and the fabrication of advanced multichip module microwave circuits. A number of heteroepitaxial structures have been reported [1–6]. Insulating materials include substituted rare earths (e.g., $\text{PrBa}_2\text{Cu}_3\text{O}_7$) [1], perovskite structured ferroelectrics [2], and dielectrics such as SrTiO_3 (STO) [3], MgO [4], and LaAlO_3 (LAO) [5]. Insulating film requirements for microwave circuit applications include low dielectric constant ($10 < \epsilon < 25$) and low loss tangents ($\tan \delta \leq 10^{-3}$). Pulsed laser deposition (PLD) is especially well suited

to the fabrication of multilayers of these complex materials since the deposition of alternating layers can be accomplished by a simple exchange of the target material. In this note, we report on the deposition and characterization of LAO on YBCO by PLD and the deposition of trilayer structures of YBCO/LAO/YBCO for passive microwave applications.

2. EXPERIMENTAL

The pulsed laser deposition system used has been described previously [7]. Briefly, the output from a Lambda Physik 315 excimer laser, operating at 248 nm, was focused with a 50-cm focal-length lens onto stoichiometric targets to an energy density of 1–2 J/cm². The evaporated material was collected onto a heated substrate positioned approximately 4 cm from the target. Films were deposited at 750°C in 300 mTorr of flowing oxygen. These conditions were

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determined to yield the best structural and electrical properties for both materials. A microprocessor controlled mirror was used to raster the laser beam across the target (0.75–2 inches in diameter) to ensure uniform deposition of thin films over the substrate. The average deposition rates were $\sim 1 \text{ \AA/pulse}$ for YBCO and $\sim 0.3 \text{ \AA/pulse}$ for the dielectrics.

The typical deposition procedure was as follows: between individual layers, the system was vented to 1 atm with oxygen and the targets were exchanged. During this time, the substrate heater setpoint was maintained at the deposition temperature. Following target exchange, the system was pumped back down to the 300 mTorr oxygen background. The targets were either sintered YBCO powder or single crystals of LAO.

Some of the dielectric layers were grown discontinuously. During the growth the deposition was interrupted by stopping the laser and raising the chamber pressure to 1 atm of oxygen. The heater temperature remained constant during this time even though the substrate surface was significantly cooler. Following the interruption, the deposition was continued at the normal deposition temperature and pressure. This interrupt procedure was done to halt the continued propagation of defect structures in the dielectric (e.g., pinholes) and promote the nucleation of new sites for film growth. For some of the device structures, patterning of the bottom superconducting layer was required. These films were removed from the deposition system, patterned, and then returned to the deposition system for the deposition of the remaining dielectric and superconducting layers.

The compositions and microstructure were characterized using Rutherford backscattering spectrometry (RBS), X-ray diffraction (XRD), and transmission electron microscopy (TEM). DC transport characteristics were measured using two techniques. When direct contact to the YBCO film could be achieved, a standard four-point probe was used to determine $R(T)$. When an insulating layer was deposited on top of the superconducting layer, T_c (and J_c) were determined inductively using a multiturn coil pressed against the film surface. Details of the inductive technique are found elsewhere [8].

Both patterned and unpatterned multilayer films were characterized at microwave frequencies. The microwave surface resistance of unpatterned superconducting films was measured by a cavity technique that has been described previously [9]. A cylindrical copper cavity, mounted on the cold stage of a closed-cycle helium refrigerator, was excited at the TE_{011}

mode at 36 GHz. The Q of the cavity was measured as a function of temperature with the YBCO film as one endwall of the copper cavity and compared to that measured with an OFHC copper endwall.

Microwave characterization of the dielectric was accomplished through the use of a microstrip transmission line which was patterned into the top superconducting layer of a trilayer structure [5]. Each of the layers was $\sim 2000\text{--}2500 \text{ \AA}$ thick. The transmission line was a 20.1-cm-long, $25\text{-}\mu\text{m}$ -wide reentrant spiral. The line had a characteristic impedance of $\sim 1 \Omega$. Because of the large impedance mismatch between the conductors in the coaxial cables and the device, the transmission line acted as a weakly coupled half-wave resonator. The transmission characteristics were measured as a function of temperature from 20 to 80 K. A complete description of this device is presented elsewhere [5].

3. RESULTS

Shown in Fig. 1 is the X-ray diffraction pattern for a multilayer structure consisting of an LAO film deposited onto a YBCO film on $\langle 100 \rangle$ MgO. The LAO was deposited at 750°C in 300 mTorr of oxygen. The diffraction pattern shows that the LAO layer is oriented on the YBCO film as evidenced by the presence of only the $(00l)$ peaks in the diffraction pattern. YBCO films were deposited onto the oriented LAO films producing a trilayer of YBCO/LAO/YBCO. Trilayer structures were deposited onto both $\langle 100 \rangle$ LAO and MgO substrates. No difference was observed in the DC or microwave transport properties of the top superconducting thin film as a function of the substrate. This is surprising since the YBCO film

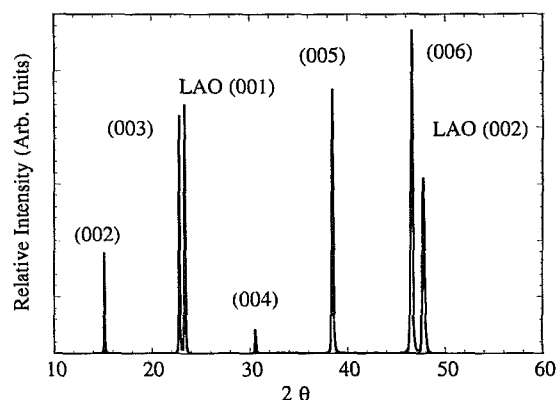


Fig. 1. X-ray diffraction scan of LAO/YBCO bilayer deposited onto $\langle 100 \rangle$ MgO.

deposited on $\langle 100 \rangle$ MgO substrates is structurally different from the YBCO film deposited on $\langle 100 \rangle$ LAO substrates. On MgO substrates, a distribution of orientations in the a - b plane is observed with maxima alignments at 0° , 45° , and 90° [10]. Only 0° and 90° alignments are observed on LAO substrates. Multilayer films (YBCO/LAO/YBCO) were deposited using shadow masks to allow examination of the individual superconducting layers. Following deposition of the first YBCO layer, one-half of the substrate was covered with a mask. The subsequent deposition of the LAO and top YBCO layers occurred only on the unmasked portion of the substrate allowing post-deposition analysis of both structural and DC transport properties of the individual YBCO layers.

X-ray diffraction analysis of YBCO films deposited at 750°C in 300 mTorr of oxygen on bilayers of LAO/YBCO indicated that the top superconducting layer was exclusively c -axis oriented. A comparison of the structural quality of the YBCO layer on a single-crystal, $\langle 100 \rangle$ MgO substrate, and the top YBCO layer of a YBCO/LAO/YBCO trilayer is shown in Fig. 2. For the bottom layer, the $(00,10)$ diffraction peak is easily resolved into two peaks which come from diffraction of the $\text{Cu } K_{\alpha 1}$ and $K_{\alpha 2}$ radiation. This is typical for YBCO films deposited on single-crystal substrates. Superimposed on this pattern is the diffraction pattern measured for the top YBCO film in a YBCO/LAO/YBCO trilayer deposited onto $\langle 100 \rangle$ MgO. The X-ray diffraction pattern is broadened slightly, which could be due to the presence of more defects, variations in film stress, and changes in oxygen stoichiometry. The peak was also shifted to a smaller 2θ , indicating a slight expansion in the c -axis

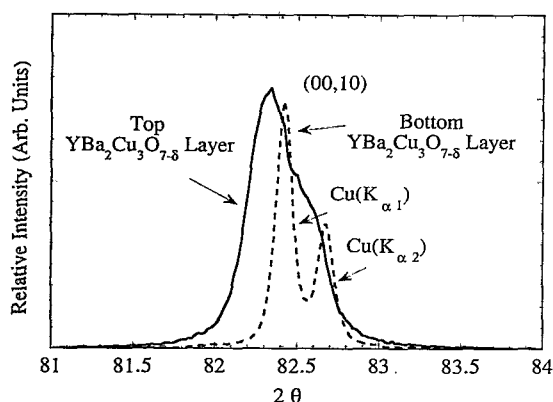


Fig. 2. X-ray diffraction pattern of the top and bottom superconducting layers in a YBCO/LAO/YBCO trilayer deposited onto $\langle 100 \rangle$ MgO.

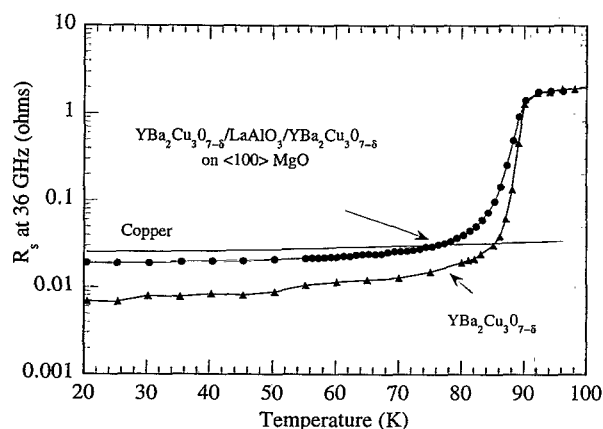


Fig. 3. Microwave surface resistance measured at 36 GHz of a YBCO film deposited onto $\langle 100 \rangle$ MgO and a YBCO/LAO/YBCO trilayer deposited onto a $\langle 100 \rangle$ LAO substrate.

lattice parameter for the top superconducting layer. The DC transport measurements also indicated small differences between the top and bottom superconducting layers in the trilayer structure. Four-point probe resistivity measurements indicate that the $R=0$ state was reduced by ~ 0.5 K. However, even in the trilayer films $R=0$ for the top superconducting layer was observed at ~ 90 K.

For microwave applications, the most important factors for multilayer thin films are the surface resistance of the superconductor and the loss characteristics of the dielectric. The microwave surface resistance of YBCO films was determined using a copper cavity/endwall replacement technique. The microwave properties of the dielectric film were obtained from patterned films.

Figure 3 shows the measured microwave surface resistance for a YBCO film deposited directly onto a single crystal $\langle 100 \rangle$ MgO substrate as well as the top superconducting layer of a YBCO/LAO/YBCO trilayer. The surface resistance of OFHC copper is also shown for comparison. The lower limit in the 36 GHz cavity R_s measurement of the YBCO films is $\sim 10\%$ of the copper background. For YBCO film on $\langle 100 \rangle$ MgO, the surface resistance shows an onset at ~ 90 K. R_s decreases sharply to ~ 85 K, then more slowly. The lowest values recorded in the 36 GHz cavity are about 3–6 mΩ at 20 K which are presumably limited by the copper background. The microwave properties of YBCO films on MgO have been characterized as well by using an all-superconducting cavity at 10 GHz [11]. The absence of the normal metal greatly increases the sensitivity of the measurement. The parallel plate resonator technique estimates the R_s to be



Fig. 4. TEM cross section of LAO/YBCO film deposited onto $\langle 100 \rangle$ MgO substrate.

$\sim 120\text{--}200\ \mu\Omega$ at 10 GHz and 20 K (which scales to $\sim 1.5\text{--}2.3\ \text{m}\Omega$ 36 GHz) [12].

The surface resistance of the top superconducting layer in a YBCO/LAO/YBCO trilayer was slightly higher than that of the YBCO film deposited directly onto $\langle 100 \rangle$ MgO, as shown in Fig. 3. Although the onset for superconducting behavior at microwave frequencies occurs at approximately the same temperature, the transition width is slightly broadened with a longer tail. The residual resistivity for the trilayer film was approximately $18\ \text{m}\Omega$ at 20 K. Extrapolation of the microwave properties to 10 GHz indicates that the top superconducting film in the trilayer structure was still better than Cu by a factor of 10 at 77 K.

Microwave losses in the trilayer transmission line were observed to be quite large (even in consideration of the large impedance mismatch) [5]. The shape of the resonances observed in the transmission line indicated the nature of the dielectric losses. If the Q of the resonant structure were constant as a function of

frequency, one would expect a broadening at higher frequencies. Instead, each resonance has approximately the same width. From circuit modeling, it was concluded that the loss mechanism was most likely due to the existence of conduction paths in the dielectric [5]. The resistivity of the dielectric was estimated at $10^6\ \Omega\ \text{cm}$.

In order to characterize the defects in the LAO, several films were examined using cross-sectional transmission electron microscopy (XTEM). Figure 4 is a TEM cross section showing the LAO/YBCO interface in a bilayer deposited onto $\langle 100 \rangle$ MgO. In the lower part of the micrograph, lattice fringes with a spacing of $\sim 12\ \text{\AA}$ are observed which indicates that the YBCO film is c -axis oriented. The LAO layer deposited onto YBCO contained a high density of threading dislocations as indicated by the arrows ($\sim 5 \times 10^{11}/\text{cm}^2$). While other defects may be present, these dislocations appear to be the dominant defect and are most likely responsible for the difference

between the thin film and the bulk conductivity. The arrows in Fig. 4 show that the threading dislocations appear to nucleate at steps on the surface of the YBCO layer. It seems likely that these dislocations serve as conduction paths between the top and bottom superconducting layers in the trilayer structure. The observation that these defects appear to nucleate at step edges in the YBCO films indicates that atomically flat YBCO films may be required for the growth of defect-free LAO films.

4. CONCLUSIONS

High-quality YBCO/LAO/YBCO structures were deposited by PLD onto $\langle 100 \rangle$ MgO and LAO substrates. These structures can be patterned into passive microwave structures. Transmission losses at microwave frequencies result from conduction currents in the dielectric. Cross-sectional TEM analysis of the multilayer films indicates a high density of threading dislocations in the dielectric layer that could be the source of the conductive paths. These defects appear to be nucleated at steps in the YBCO layer. Techniques that can reduce the surface roughness on

an atomic scale may be required to minimize the defect density in the dielectric layer.

REFERENCES

1. C. T. Rogers, A. Inam, M. S. Hedge, B. Dutta, X. D. Wu, and T. Venkatesan, *Appl. Phys. Lett.* **55**, 2032 (1989).
2. Y. A. Boikov, S. K. Esayan, Z. G. Ivanov, G. Brorsson, T. Claeson, J. Lee, and A. Safari, *Appl. Phys. Lett.* **61**, 528 (1992).
3. J. J. Kingston, F. C. Wellstood, P. Lerch, A. H. Miklich, and J. Clark, *Appl. Phys. Lett.* **56**, 189 (1990).
4. S. Tanaka, H. Nakanishi, T. Matura, K. Higaki, H. Itozaki, S. Yazu, *IEEE Trans. on Mag.* **27**, 1607 (1991).
5. J. M. Pond, K. R. Carroll, J. S. Horwitz, D. B. Chrisey, M. S. Osofsky, and V. C. Cestone, *Appl. Phys. Lett.* **59**, 3033 (1991).
6. Y. A. Boikov, G. Brorsson, T. Claeson, and Z. G. Ivanov, *Appl. Phys. Lett.* **59**, 2602 (1991).
7. D. B. Chrisey, J. S. Horwitz, H. S. Newman, M. E. Reeves, B. D. Weaver, K. S. Grabowski, and G. P. Summers, *J. Superconductivity* **4**, 57 (1991).
8. J. H. Claussen, M. E. Reeves and R. J. Soulen, *Rev. Sci. Instrum.* **64**, 996 (1991).
9. H. S. Newman, D. B. Chrisey, J. S. Horwitz, B. D. Weaver, and M. E. Reeves, *IEEE Trans. on Magnetics* **27**, 2450 (1991).
10. B. H. Moeckly, D. K. Lathrop, S. E. Russek, R. A. Buhrman, M. G. Norton, and C. B. Carter, *IEEE Trans. on Magnetics* **27**, 1017 (1991).
11. R. C. Taber, *Rev. Sci. Instrum.* **61**, 2200 (1990).
12. R. C. Taber, private communication.